Seasonal Variation in the Difference between Observed and Calculated Particulate Fluxes of Th-234 in Funka Bay, Japan

Shizuo Tsunogai†, Kazunori Taguchi† and Koh Harada†

Abstract: Particulate fluxes were determined by two methods to elucidate the behavior of settling particles in seawater. One method involves direct observation of fluxes with sediment traps, while in the other method flux is indirectly calculated from the radioactive disequilibrium between U-238 and Th-234 in seawater, which gives net flux. Observations were carried out several times throughout a year in Funka Bay. When linearly extrapolated, the observed gross fluxes of Th-234 did not converge to zero at the surface. In the subsurface water the difference between the observed and calculated fluxes showed a seasonal variation. The observed fluxes roughly coincided with the calculated net fluxes in the summer stratified water but the observed fluxes were much larger than the calculated ones in the convective winter water. Conversely the observed fluxes were smaller than the calculated ones in spring when the water was exchanging. These results suggest that we can apply this two method to get information not only on the behavior of settling particles in seawater but also on the physical stability of water.

1. Introduction

Recently sediment traps have popularly been used to reveal various phenomena occurring in the ocean and many valuable results have been obtained through sediment trap experiments (Wiebe, 1976; Honjo, 1978, 1982; Knauer et al., 1979; Brewer et al., 1980, Iseki, 1981; Deuser et al., 1981, Tsunogai et al., 1982; Bacon et al., 1985). There are, however, some unsolved technical problems still remaining. Among them the most important one is the trapping efficiency of the sediment trap (Soutar et al., 1977; Hargrave and Burns, 1979; Gardner, 1980ab, 1985; Blomqvist and Kofoid, 1981; Bruland et al., 1981; Dymond et al., 1981; Staresinic, 1982). It is extremely difficult to reach a final conclusion about the problem because settling particles are an assemblage of various particles that differ in shape, size, density, composition, etc., and because the physical conditions of each water column where the sediment trap is deployed differ from observation to observation.

Gardner (1980b) has stated that a cylinder with a height/width ratio of 2.3 yields an accurate vertical flux while traps with larger height/width ratios overtrap fine particles, although tilt effects have also been considered (Gardner, 1985). Sato (1981) has observed that all the cylinders with height/width ratios larger than 3 give similar downward fluxes in Mikawa Bay and concluded that the flux observed by the longer cylinders can be regarded as a true vertical flux.

Aside from the trapping efficiency problem, Tsunogai et al. (1980) have discussed the trapping rate from an alternative aspect of the settling process. They attributed the enormous downward particulate flux observed in winter in Funka Bay to the behavior of settling particles in the physically unstable water. Furthermore Tsunogai et al. (1982) have used the same reasoning to explain the fact that the vertical flux of aluminum increases with depth in the ocean. They suggested that a vertical advective (up and down) motion of the physically unstable water increases the apparent particulate flux and the advective motion is related to the static stability, namely the vertical density gradient of the water. To confirm this reasoning we have compared the observed particulate fluxes with net fluxes calculated from the radioactive disequilibrium between U-238 and Th-234 in seawater.

The concentration of Th-234 (half-life, 24 days)
in seawater is balanced by production from its parent U-238 (half-life, $4.5 \times 10^9$ yrs) dissolved in seawater by making a stable complex anion and loss due to its radioactive decay and various removal processes. Therefore the net rate of removal of thorium can be calculated from the concentrations of U-238 and Th-234 in seawater if the physical water movement is known and modelled.

The hydrography of Funka Bay has been studied well and a closed system model can be practically applied to the bay water except during the two water mass exchange periods in spring and autumn (Ohtani, 1981, a review). The model has already been applied successfully to determine the removal rate of Th-234 in Funka Bay water (Minagawa and Tsunogai, 1980; Tanaka et al., 1983).

2. Methods

The observations were carried out at a station (42°16.6'N, 140°35.0'E; 92 m depth) in Funka Bay (Fig. 1) from April 1981 to March 1982.

Seawater samples were collected eight times from each of ten depths at intervals of 10 m from the surface to 90 m by pumping. For pumping, a submersible pump (Teikoku Denki, YH-110) and a 25 mm plastic tube were used. The pumping rate was about $201 \text{ min}^{-1}$. The seawater samples were analyzed for Th-234 as well as salinity and other natural radionuclides which will be reported elsewhere. The analy-

![Fig. 1. Sampling station in Funka Bay. The station is shown with a solid circle.](image)

![Fig. 2. Vertical profiles of concentration of Th-234 in dpm l$^{-1}$ in Funka Bay water. The sampling date is also written near each profile. Short bars along with U-238 refer to the mean concentration of U-238 in units of radioactivity (dpm l$^{-1}$).](image)
tical method for Th-234 in seawater was the same as that of Harada and Tsunogai (1985).

The sediment traps (U-type) were deployed seven times at each of three depths, usually 1 or 10, 40 and 80 m. The dimensions of the cylinder type sediment trap was 7 cm in diameter and 60 cm long. The traps were so long that lids were unnecessary, although we attached lids to traps deployed after December 1981. The mooring system was almost the same as that of Tsunogai et al. (1980). The sediment traps were tightly dragged with subsurface buoys except that the surface trap was lowered from the surface by a lead. After recovering the traps which were deployed for 3 to 14 days, as much supernatant water as possible was syphoned off and the trapped particulate matter was collected on filter paper. Dry weight of the filters was measured by the method of Uematsu et al. (1978). The particles were dissolved with a mixture of concentrated HNO₃, HClO₄ and HF. The subsequent procedure for the Th-234 determination was the same as that for Th-234 in seawater.

3. Results and discussion

3.1. Seasonal and vertical variation of Th-234 in seawater

The concentration of Th-234 in seawater is shown in Fig. 2. The observed results for Th-234 in seawater in 1981-1982 show a similar tendency to those that Tanaka et al. (1983) observed at the same station in 1979-1980, although they did not observe the spring bloom of phytoplankton in March. The seasonal change in the Th-234 concentration in seawater is more clearly demonstrated in this work.

In summer the concentrations of Th-234 were...

<table>
<thead>
<tr>
<th>Date</th>
<th>Depth (m)</th>
<th>Total flux (g m⁻² day⁻¹)</th>
<th>Th-234 Concentration (dpm g⁻¹)</th>
<th>Flux (Fobs) (dpm m⁻² day⁻¹)</th>
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* Concentrations at the retrieval are shown and those corrected for the radioactive decay during the deployment are also shown in parentheses. The error due to counting statistics corresponds to the 1σ value of each count.
smaller in the surface and bottom waters. Thus, the maximum concentrations were found in the mid-layer between 40 and 60 m in the summer stratified water. This should be due to “regeneration” of Th-234 from settling particles. However, neither a maximum of nutrient concentration nor a minimum of oxygen content has been observed in the mid layer. Tsunogai and Minagawa (1978), therefore, considered that this phenomenon is due to decrease in the mean settling velocity of particulate matter due to break down to fine particles having slower sinking rates in the stratified water.

In winter active convection of water induced by cooling at the surface homogenized the vertical concentration of Th-234. From winter to spring when the spring bloom of phytoplankton came (Tsunogai and Watanabe, 1983), the concentration of Th-234 decreased rapidly in the surface layer. Biological production of settling particles is a major sink of Th-234 in seawater.

3.2. Seasonal variation of observed particulate fluxes

The results obtained in the sediment trap experiments are shown in Table 1. The total fluxes of Th-234 ($F_{obs}$) are based on the radioactivities of Th-234 at the time of recovery of the sediment traps which are not corrected for decay during the deployment at sea. Correction can be made if the deployment interval is sufficiently short (a few days) or if the daily flux does not change with time. The corrected daily mean flux ($\bar{F}_{obs}$) assuming a constant value is given by

$$\bar{F}_{obs} = \lambda \Sigma F_{obs} / (1 - e^{-\lambda T}) \quad (1)$$

where $T$ is the time interval between the deployment and the retrieval of the sediment traps.

The daily mean fluxes ($\bar{F}_{obs}$) given in Table 1 indicate extremely large values in the convective winter water and smaller values in the stratified summer water. This seasonal variation coincides with that obtained and discussed by Tsunogai et al. (1980), although they did not carry out the observations during the spring bloom of phytoplankton in March. The flux of Th-234 in late February to March was high compared with that in summer, and this results in a decrease in the concentration of Th-234 in seawater in March.

3.3. Calculation of net downward flux of Th-234 in the water column

As shown in section 3.1., a steady state model does not apply to Th-234 in Funka Bay water. According to Tanaka et al. (1983), the change in the inventories of Th-234 in a closed system in a non-steady state is expressed by the following equation;

$$A_2 = \frac{\lambda A_p}{(\lambda + k)} + \left[ A_1 - \frac{\lambda A_p}{(\lambda + k)} \right] e^{-(\lambda + k)T} \quad (2)$$

where $A_1$ and $A_2$ are the inventories of Th-234 at the initial and the last observation in units of radioactivity (dpm/m²), $A_p$ is the inventory of its parent, U-238, in radioactivity assuming a constant concentration with time, $\lambda$ is the decay constant of Th-234 (1/35 day⁻¹), $T$ is the observation interval and $k$ is a removal rate constant assuming a first-order removal process.

The removal rate constant, $k$, is calculated from Eq. (2) or the following Eq. (3) if the observation interval is sufficiently short, say, less than one week;

$$k \doteq \lambda [A_{p2}/A_1 - 1] + [1 - A_{p2}/A_1]/T. \quad (3)$$

The residence time of thorium thoroughly mixed with Th-234 or that of Th-234 excluding a sink due to its radioactive decay is given by

$$\tau = 1/k \quad (4)$$

Fig. 3. Residence time (τ) of Th-234 in Funka Bay in days calculated from Eqs. 2-4. Solid lines indicate the values obtained in this work and broken lines indicate those obtained in 1979-1980 by Tanaka et al. (1983).
### Table 2. Salinity and concentration and inventory of Th-234 in Funka Bay water.

<table>
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<tr>
<th>Depth (m)</th>
<th>Salinity (%)</th>
<th>Th-234 Concentration (dpm m⁻³)</th>
<th>Inventorya (10⁵dpm m⁻²)</th>
<th>Depth (m)</th>
<th>Salinity (%)</th>
<th>Th-234 Concentration (dpm m⁻³)</th>
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<td>113.6</td>
<td>60</td>
<td>33.507</td>
<td>1.08±0.03</td>
<td>44.2</td>
</tr>
<tr>
<td>70</td>
<td>33.059</td>
<td>2.08±0.09</td>
<td>133.8</td>
<td>70</td>
<td>33.519</td>
<td>0.99±0.02</td>
<td>54.5</td>
</tr>
<tr>
<td>80</td>
<td>33.326</td>
<td>1.68±0.08</td>
<td>152.6</td>
<td>80</td>
<td>33.529</td>
<td>0.89±0.02</td>
<td>63.9</td>
</tr>
<tr>
<td>90</td>
<td>33.705</td>
<td>0.87±0.06</td>
<td>165.3</td>
<td>90</td>
<td>33.559</td>
<td>1.57±0.06</td>
<td>76.2</td>
</tr>
</tbody>
</table>

*a* Amounts above the level quoted.  
*b* Interpolated values.
where $\tau$ is the residence time. In the calculation the concentration of U-238 is assumed to be proportional to salinity and to be 2.45 dpm l$^{-1}$ for 35% of salinity.

The calculated residence times of thorium in a water column of 90 m (Fig. 3) shows good agreement with those obtained at the same station in 1979–1980 by Tanaka et al. (1983). The residence time during spring bloom of phytoplankton in March was found to be extremely short at about two weeks, indicating the importance of biological activity in the removal of thorium, a highly insoluble metal, from seawater.

The net flux of Th-234 is simply calculated from the change in the inventories of Th-234 in seawater independent of the removal process, namely without assuming a first-order rate constant as follows;

$$\Sigma F_{\text{calc}} = A_0 (1 - e^{-\lambda \tau}) + A_1 e^{-\lambda \tau} - A_2 \quad (5)$$

where $\Sigma F_{\text{calc}}$ is the amount of Th-234 to be trapped after deployment for $T$ days, $A_1$ and $A_2$ are the inventories of Th-234 at the deployment and retrieval dates, respectively, in the water column above the sediment trap deployed. The three terms on the right side are the amount of Th-234 grown from U-238 during the deployment, original Th-234 remaining at the retrieval and the amount of Th-234 existing in the water column at retrieval.

Unfortunately the above equation can only be applied to the last set of observations on 1 and 15 March 1982 (Table 1 and Fig. 2), because the dates for sampling seawater were somewhat different from those for the sediment trap experiments. Then for these cases we are forced to assume a first-order removal process to obtain $A_1$ and $A_2$ values in Eq. (5), which are the inventories of Th-234 at the deployment and the retrieval dates of the corresponding sediment trap experiment, by inserting the
formerly obtained value into Eq. (2). In this
case the daily flux of Th-234 in a non-steady
state condition \( \overline{F}_{\text{cale}} \) is calculated from the
following equation:

\[
\overline{F}_{\text{cale}} = \lambda \int_0^T kN \, dt / T \\
= \lambda k A_p / (\lambda + k) + k \left[ A_1 - \lambda A_p / (\lambda + k) \right] \\
\times \left[ 1 - e^{-(\lambda + k)T} \right] / T (\lambda + k)
\]

(6)

where \( N \) is the number of atoms of Th-234
(i.e., the inventory of Th-234) and \( T \) is the
duration of the sediment trap experiment. The
time is set to zero and the inventory of Th-234
in units of radioactivity is \( A_1 \) at the time of
deployment of the sediment trap.

The calculated daily fluxes \( \overline{F}_{\text{cale}} \) are obtained
from the data given in Table 2 and compared with
the observed daily fluxes \( \overline{F}_{\text{obs}} \) in Fig. 4. The observed flux \( \overline{F}_{\text{obs}} \) is always larger than
the calculated net flux \( \overline{F}_{\text{cale}} \) in the layer near
the surface. That is to say, the observed flux
\( \overline{F}_{\text{obs}} \) at the surface obtained by extrapolation
is not zero, but it should be zero if Th-234
atoms produced in the water column were
removed from the water at a constant rate.
Further the difference between \( \overline{F}_{\text{obs}} \) and \( \overline{F}_{\text{cale}} \)
at the surface is much larger in winter. In
other words, the difference between the two
particulate fluxes is not proportional to the net
flux but changes from season to season. These
results cannot be explained by mere over-trapping
due to artifacts, and the results indicate existence
of the “upward” flux defined by Tsunogai et al.
(1980).

We have found a distinct seasonality in the
difference between \( \overline{F}_{\text{obs}} \) and \( \overline{F}_{\text{cale}} \) in the
subsurface layer too (Fig. 4). The observed gross
flux \( \overline{F}_{\text{obs}} \) agrees fairly well with the calculated
net flux \( \overline{F}_{\text{cale}} \) in the stratified summer water,
but the gross flux is much larger than the net
flux in the unstable winter water. This con-
firms the conclusion obtained by Tsunogai et al.
(1980) that the physical stability of the water
has a large affect on particulate flux observed
by the sediment traps.

The calculated net flux during the spring
bloom of phytoplankton in March is larger than
the gross flux observed with sediment traps.
Although the reason for this is uncertain, the
closed system model may not apply to the spring
water because Oyashio water flowed to the Bay
at that time.

Because of its short half-life, Th-234 is useful
for calculating the net flux in the surface water.
When we study this problem in the deep ocean,
Pb-210 (half-life, 22 yrs) and Th-230 (half-life,
75,000 yrs) will be useful tracers. In our future
work we aim to study quantitatively the difference
between the observed gross and the calculated
net fluxes by using these nuclides. The results
will also give information on the physical motion
of water masses.

How shall we treat the results obtained with
sediment traps when the observed flux is signifi-
cantly different from the calculated flux? At
present we cannot explicitly answer this question
because the correction using the calculated flux
is not always applicable to a component contained
in settling particles that differ from those con-
taining the radionuclides. We can only say,
“Pay attention not only to absolute fluxes but
also to the relative concentration observed with
sediment traps”.

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Seasonality in the supply of sediment to the deep
Sargasso Sea and implications for the rapid
Tsunogai, Taguchi and Harada


噴火湾における Th-234 の粒子束の実測値と計算値との差の季節的変動

角皆靜男*, 田口和典*, 原田晃*

要旨: 海水中の粒子の挙動を知るために、2つの方法で粒子束を決定して比較することを考えた。セミメートトラップで実測する方法と Th-234 の放射能平衡を利用する方法である。観測は噴火湾で1年中年にわたり数回行なった。表面での Th-234 粒子束の実測値は 0 に取東せず、また冬に大きな値を示した。また表面下の混層でも実測値と計算値の間の差は季節的に変動し、夏の显存量に小さく、冬の混層期にきわめて大きかった。水が交替する春には逆に計算値の方が大きかった。この比較法は海水中の粒子の挙動ばかりではなく、水の物理的安定性に関する情報をも与える可能性があることがわかった。

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